CONTINUOUS RECYCLING IN THE HOMOGENIZATION OF RELATIVELY SMALL SAMPLES

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SUMMARY

The mathematical relationships governing continuous recycling in the homogenization of small samples have been derived and compared with experimentally determined relationships. Distribution curves have been obtained showing the fraction of milk at any time t which has undergone p homogenizations. An empirical equation has been derived, showing the relationship in intermittent recycling between the degree of homogenization as measured by turbidity, and the number of homogenizations. Combining the equations defining the distribution curve with the empirical equation permits the calculation of the degree of homogenization at any time t of a sample undergoing continuous recycling in homogenization.

Posing the question of the effect of multiple passes through a homogenizer, Deackoff and Rees (1) showed conclusively that with respect to the homogenization index which they employed, the effect of increasing the number of passes was tantamount to increasing homogenization pressure.

This effect may have interesting application in the homogenization of small samples, 3–10 lb. of milk, with a 125-gal.-per-hour plant-size homogenizer. Such small samples are usually lost because of the time spent in adjusting to pressure at the beginning of an operation, and because of the loss of pressure at the end.

If time is considered as a variable in homogenizing small samples, and if the homogenized portions are fed back continuously to the sample, it becomes possible to achieve degrees of homogenization resembling those obtained at pressures beyond the pressure range of the homogenizer. Aside from a small uncertainty involved in bringing duplicate samples to the same homogenization pressures, the procedure allows for a much higher degree of reproducibility than that which is obtained with normal homogenization techniques. However, in recycling continuously there will always be in theory a diminishingly small portion of sample which has not undergone homogenization and, by the same token, there will always be an infinitesimal portion which has undergone an infinite number of homogenizations.

It is the purpose of this paper to derive the necessary mathematical relationships governing continuous multiple-pass homogenization (hereinafter designated as multipass homogenization), and to compare the data deduced from theory with those obtained in experiment.

THEORY

In the mathematical formulation, the following symbols are employed:

V_o = Volume of sample in gallons.

V_t = Volume passing through homogenizer in time interval t.

ΔV, Δf = Increment of volume and fractional increment of volume, respectively, passing homogenizer valve in time interval Δt.

n = Number of increments of volume.

$f_{n, p}$ = Fraction of total volume which has undergone p homogenizations following the passage of n increments of volume.

$V_{n, p}$ = Volume which has undergone p homogenizations following the passage of n increments of volume.

C = Capacity of homogenizer in gallons per minute.

m = Number of times volume V_o is recycled.

k_p = Turbidity coefficient after p homogenizations.

K = Turbidity of sample at time, t.

Consider an ideal arrangement in which an increment of volume ΔV is removed from the sample and, following the homogenization of this small sample, is added back to the main sample. As a result, the sample will be found to contain a portion, $V_o\left(1 - \dfrac{\Delta V}{V_o}\right)$ gal. with a turbidity coefficient k_o, and a portion, ΔV gal., with a turbidity coefficient k_1. If these operations are repeated n times it is easily shown that the following relationships apply:

(1) $f_{n, 0} = (1 - \Delta f)^{n}$; $n \geqq 0$

(2) $f_{n, 1} = n \Delta f (1 - \Delta f)^{n-1}$; $n \geqq 1$

(3) $f_{n, 2} = \dfrac{n (n-1)}{2 !} (\Delta f)^2 (1 - \Delta f)^{n-2}$; $n \geqq 2$

(4) $f_{n, p} = \dfrac{n !}{(n-p) ! \, p !} (\Delta f)^p (1 - \Delta f)^{n-p}$; $n \geqq p$

(5) $\Delta f = \dfrac{m}{n}$; substituting in (4)

(6) $f_{n, p} = \dfrac{n !}{(n-p) ! \, p !} \left(\dfrac{m}{n}\right)^p \left(1 - \dfrac{m}{n}\right)^{n-p}$

Consider the system after a large number of increments have been homogenized. It follows that for all values of p and for $m = \dfrac{Ct}{V_o} > 0$, the values of $f_{\infty, p}$ are given by the equation:

(7) $f_p = \dfrac{\left(\dfrac{Ct}{V_o}\right)^p e^{-\frac{Ct}{V_o}}}{p!}$

Equation (7) shows the relationship between that fraction of a sample which has undergone p homogenizations and time t of homogenization. The equations show quite clearly that, if the volume of samples and the time of homogenization are multiplied by the same factor, the same size distribution should prevail at corresponding times. This relationship suggests one means for testing Equation (7).

We shall now, utilizing Equation (7), obtain a relationship between turbidity and homogenization time. The turbidity of a turbid solution or suspension is defined thus:

$$(8)\quad K = \frac{1}{l} \log \frac{I_o}{I}$$

Where l, I_o, and I are, respectively, the length of the light path, the incident light intensity, and the attenuated light intensity. The value of K, which is a function of particle size and shape, the refractive index of the solvent, the refractive index of scattering units of the same kind, and the wave length of the light employed in the measurement, may be deduced from theoretical considerations based on electromagnetic theory. Leviton (2) has applied the Mie theory to a study of the effect of the colloidal constituents of milk on light scattering. However, in this paper, the Mie theory will not be considered. Rather, calculation of turbidity values will be based in part on Equation (7), and in part on the empirical relationship prevailing between turbidity and the number of passes through a homogenizer valve which a sample undergoes in an intermittent multipass system. If k_o k_1, k_2, ... k_p are turbidity coefficients corresponding to 0, 1, 2, ... p passes, then the turbidity of a sample in a continuous multipass arrangement is given by the relationship:

$$(9)\quad K = \log \frac{I_o}{I} = \sum_{p=o}^{p=\infty} f_p\, k_p$$

Measurements, it is assumed, are made with a Model B Beckman spectrophotometer with 1-cm. absorption cells at 1,020 mμ, according to the procedure of Deackoff and Rees (1). The unit of concentration, accordingly, is the concentration (grams per milliliter) of fat in a suspension containing 1 ml. milk (3.5% fat), 5 ml. of 5 N ammonia, and 250 ml. H$_2$O, and K, and the various k's refer to such a solution. If K is the turbidity of a solution containing more or less than this amount of fat, then $K = \dfrac{3.5\,K'}{\text{fat concentration in milk in per cent}}$.

The value of k_o depends on the size distribution of the fat globules in the original milk. The values of k_1, k_2, etc., depend on this size distribution, on the composition of the milk and, more important, on the homogenization temperature and pressure, and on the properties of the homogenizer valve.

EXPERIMENTAL PROCEDURE

Milk from a mixed herd was standardized to contain 3.5% fat. It was pasteurized at 63° C. for 30 min. and cooled. A 25-lb. sample was warmed to 75° C. and fed into a reservoir. This reservoir was connected to the inlet of the homogenizer by means of minimum lengths of sanitary tubing of approximately $\frac{7}{8}$-in. diameter. In recycling experiments, the outlet was also connected to the reservoir with sanitary tubing. To remove frictional heat, a portion of the return tubing was wrapped in toweling over which water was allowed to drip.

In the first series of experiments, the values of k_o, k_1, ... k_8 were determined. The milk was passed through the homogenizer eight times and a sample for turbidity measurement was taken after each pass. The temperature and the homogenization pressure (500 lb. per square inch on the second stage and 3,000 lb. per square inch on the first) were adjusted before each pass.

Five- and ten-pound samples were next homogenized continuously. The connecting tubing was warmed and the milk wash allowed to circulate for approximately 15 sec. before the application of 500 lb. per square inch on the second-stage valve. The first-stage valve was then adjusted, timing begun, and cooling water applied. Samples of approximately 10 ml. were withdrawn at 30- and 60-sec. intervals, respectively, in the experiments in which 5 and 10 lb. of milk were employed. The smallest sample which could be recycled conveniently weighed approximately 3 lb. With this small sample the swirling motion in the reservoir was damped by the use of a cross-shaped bar over the orifice.

<center>RESULTS AND DISCUSSION</center>

Table 1 shows the results of turbidity measurements on samples which have been homogenized from zero to eight times. The transmittance values given in the last column are those calculated from empirical equation:

$$(10) \quad k_p = \log \frac{I_o}{I} = (k_o - k_\infty)\, e^{-a\sqrt{p}} + k_\infty$$

The values of k_∞ and a depend largely on the conditions of homogenization; k_∞ is the turbidity corresponding to an infinite number of passes, and k_o is the turbidity of the unhomogenized milk. It is quite likely that the value of k_∞ becomes nearly independent of pressure in the high-pressure range, inasmuch as k_1 appears to become nearly independent of pressure at high pressure (1). The value for k_∞ (given in Table 1) is that obtained from turbidity measurement

<center>TABLE 1</center>
<center>Comparison of observed and calculated transmittances for milks undergoing
intermittent multipass homogenization</center>

Passes (p)	Turbidity (k_p) (observed)	Transmittance I/I_o (observed)	I/I_o (calculated)
		(%)	
0	0.362	43.3	43.3[a]
1	0.136	73.0	71.5
2	0.102	78.7	78.0
3	0.084	82.5	81.5
4	0.077	83.8	83.7
5	0.069	85.2	84.8
6	0.063	86.0	86.0
7	0.060	86.8	86.8
8	0.057	87.6	87.4
∞	0.046	90.1	90.1[a]

Calculated values obtained from empirical equation:

$$k_p = \log \frac{I_o}{I} = (k_o - k_\infty)\, e^{-a\sqrt{p}} + k_\infty = 0.3159\, e^{-1.16\sqrt{p}} + 0.0461.$$

[a] Values taken to be the same as those observed.

of a sample of milk which in continuous multipass homogenization had under-gone the equivalent of 45 homogenizations. The empirical relationship furnished a basis for extrapolation of the experimentally determined values to include many intractable to direct measurement. This extrapolation is necessary to test the validity of Equation (9).

Figure 1 is a plot of Equation (7) for values of $m = \dfrac{Ct}{V_o}$ ranging from 1.91 to 13.37. The resemblance of the family of curves to those which are met in
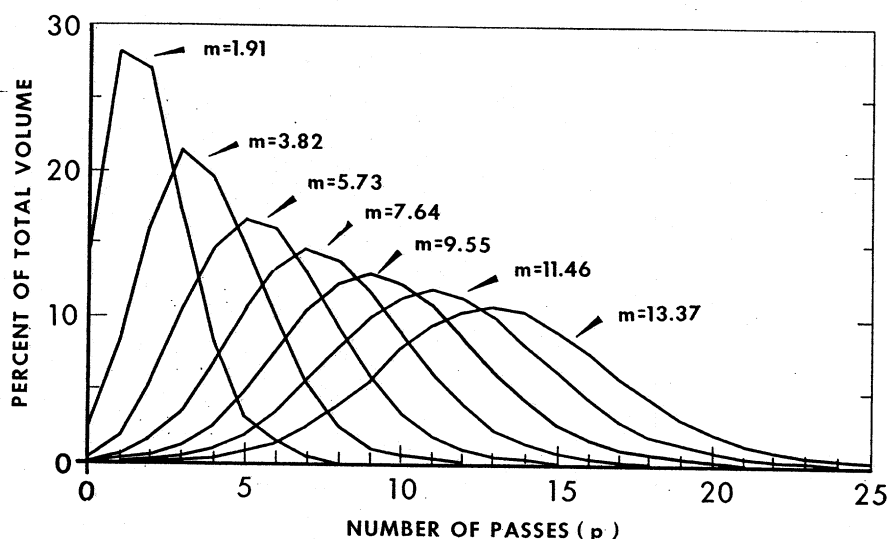

FIG. 1. The percentage of the total volume which has undergone p homogenizations in con-tinuous multipass homogenization is shown for various values of m.

probability theory is no accident, inasmuch as Equation (6), from which Equation (7) derives, lends itself to formulation in terms of probability theory. Thus, $f_{n,\,p}$ is the probability that as n approaches infinity a fat globule selected at random will have undergone p homogenizations. The maximum value of f_p occurs at an integral value of p for which $m > p > m - 1$. The mean value of f_p is equal to m and the standard deviation is equal to $\sqrt{m}$. Thus, the dispersion in-creases, that is, the value of the maximum decreases and the standard deviation increases as the time of homogenization increases.

Equation (9) shows that the resultant turbidity of a sample in continuous multipass homogenization is the weighted sum of the turbidities in intermittent multipass homogenization, taken over the range of passes from zero to infinity. The weighting factors are the f_p's derived from Equation (7), and turbidity as a function of the number of passes is given by Equation (10).

In Figure 2, the turbidities calculated by means of Equations (9) and (10) are compared with experimental values. Good agreement is obtained over prac-tically the entire range of abscissas under survey. Some uncertainty exists in the selection of a zero time and, hence, it is to be expected that the percentage
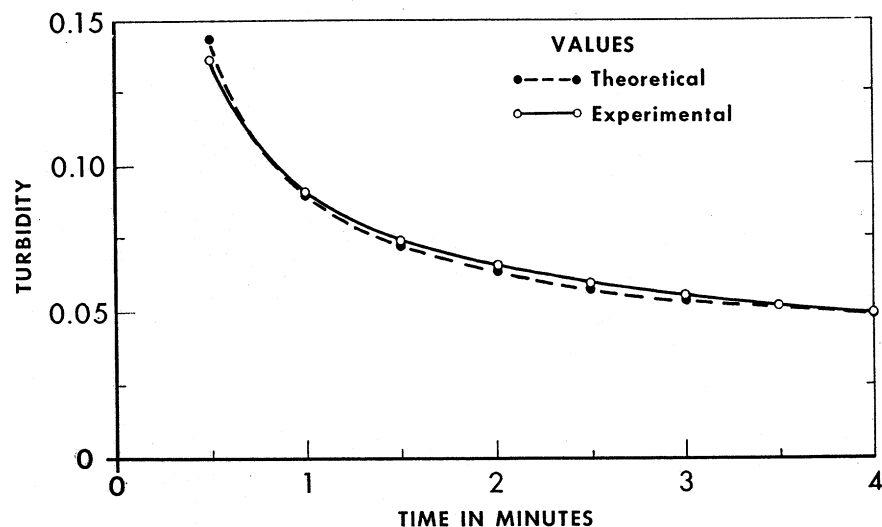
Fig. 2. Experimental and calculated values for the turbidity of homogenized milk are shown as a function of the time of homogenization. (Volume of sample, 0.55 gal.)

error will be greatest in the early stages of homogenization. Furthermore, the conditions underlying the development of Equation (7), i.e., no holdup of sample before and after homogenization, are not followed strictly. We have assumed perfect mixing of the milk in the reservoir and in the conduit space leading to the homogenization valve, and no allowance has been made for the effect of holdup (approximately 0.5 lb.) in the return piping. The value of V_o has been taken equal to the volume of milk processed, minus the volume held up in the return piping. Holdup in the return piping, it is reasonable to expect, would reduce homogenization efficiency slightly.

As a further check on the validity of Equations (7), (9), and (10), a comparison was made of the time required to effect the same degree of homogenization in two samples differing in volume. According to Equation (7), the time should vary directly as the volume and, hence:

$$\frac{Ct_1}{V_1} = \frac{Ct_2}{V_2} = \text{constant.}$$

The results (given in Table 2) tend to support this conclusion.

In the assignment of values to the constants in Equation (11), only the selection of the value for a is based on the method of least squares; k_o is the turbidity of the unhomogenized sample and, hence, is independent of processing conditions; k_∞ is a maximum turbidity value, and its measurement is not subject to the relatively high degree of error associated with the measurement of turbidity in the intermediate range.

The relationships which have been developed may be used in a converse sense, that is to say, from measurements of turbidity in a continuous multipass system, the constants of Equation (10) may be calculated and, hence, the tur-

TABLE 2

Table shows that the time required for a sample to attain a certain degree of
homogenization is proportioned to the volume of sample

Time (t)	Turbidity	$\left(\dfrac{Ct}{V_o}\right)$ Calculated[a]	$\left(\dfrac{Ct}{V_o}\right)$ Measured[b]
(min.)			
1	0.131	2.15	1.81
2	0.092	3.80	3.62
3	0.077	5.10	5.43
4	0.064	7.00	7.24
5	0.060	9.00	9.05
6	0.056	10.50	10.86
7	0.053	12.30	12.67

[a] Calculated from data in Figure 2 and relationship: $\dfrac{Ct}{V_o} = \text{constant}$.

[b] $C = 2.1$ gal. per minute. $V_o = 1.16$ gal.

bidities belonging to samples in intermittent multipass homogenization may be
calculated as a function of the number of passes. This procedure has elegance
and simplicity to recommend it, compared with direct measurement.

Although the fractional distribution of the number of homogenizations is such
that the mean number is equal to the number of times the total volume has been
cycled, it does not follow *a priori* that the turbidity of a sample which has been
cycled m times is equal to the turbidity of a sample which has undergone m
homogenizations. The results given in Figure 3 represent an attempt to settle
the matter experimentally. They show that following the first two passes the
increase in turbidity per cycle in continuous homogenization is approximately
equal to the increase per pass in intermittent homogenization. Actually, the curve
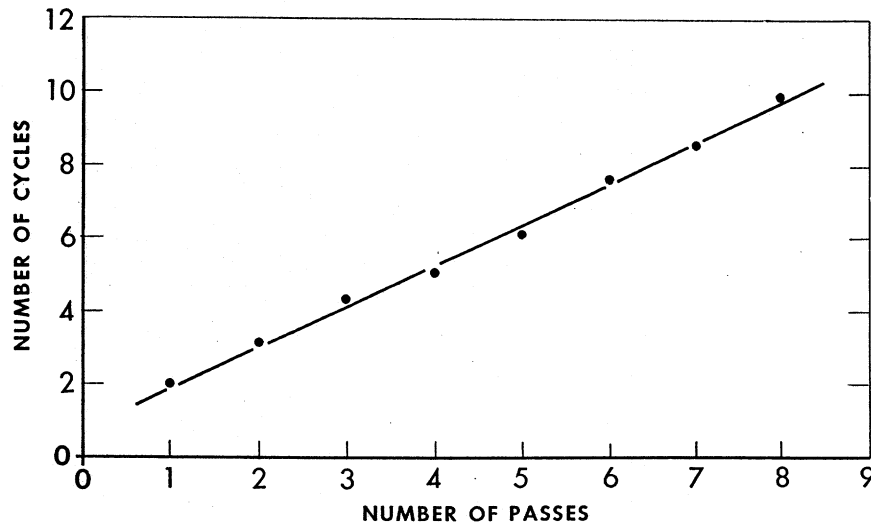

FIG. 3. The number of times a sample must be cycled in continuous multipass homogeniza-
tion, to achieve definite degrees of dispersion, is shown as a function of the number of passes
required in intermittent homogenization to achieve the same results.

indicates that 1.1 cycles is equivalent to one pass. The approximate relationship is clearly indicated in Equations (9) and (10) only for very large values of m.

The degree of homogenization as measured by turbidity does not determine uniquely the degree of dispersion of the fat phase. Whether milk in which a high homogenization pressure has been employed, to achieve a certain high degree of homogenization, has the same degree of dispersion as milk in which the same high degree of homogenization has been obtained at lower pressures in a multipass arrangement, is a matter best settled by particle size measurement; or, if the system does not lend itself to direct measurement, measurement of some property which is sensitive to differences in size distribution can be made.

ACKNOWLEDGMENT

In a careful and constructive review by the Editorial Board of this Journal an alternate derivation of Equation (7) based on probability theory was suggested. In acknowledgment, this derivation is presented herewith:

The total number of fractions treated after m cycles is mn. The probability of p successes in mn trials is given by the formula (available in mathematical handbooks)

$$f_p = \frac{(mn)!}{p!\,(mn-p)!} \left(\frac{1}{n}\right)^p \left(1 - \frac{1}{n}\right)^{mn-p}$$

This is directly applicable to find the probability of a particular drop of milk that has been homogenized p times. Stirling's approximations of factorials shows that

$$n! = \sqrt{2\,\pi\,n}\ \left(\frac{n}{e}\right)^n.$$

Substituting into the equation for f_p and simplifying, one obtains:

$$f_p = \frac{m^p}{p!\,e^p} \left(1 + \frac{p-m}{mn-p}\right)^{mn-p}$$

or

$$f_p = \frac{m^p}{p!\,e^p} \left[\left(1 + \frac{1}{\frac{mn-p}{p-m}}\right)^{\frac{mn-p}{p-m}} \right]^{p-m}$$

When n approaches infinity, the process becomes a truly continuous recycling homogenization and the equation becomes:

$$f_p = \frac{m^p}{p!\,e^p} e^{p-m} = \frac{m^p}{p!} e^{-m}$$

which is Equation (7).

REFERENCES

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